

IN THE UNITED STATES PATENTS AND TRADEMARK OFFICE

Appln No. : 10/568,967
Confirm. No.: 8880
Applicant(s): Hideo Taka et al.
Filed : March 14, 2006
For : ORGANIC ELECTROLUMINESCENT...
Art Unit : 1794
Examiner : MICHAEL H. WILSON
Docket No. : 06096/HG
Customer No.: 01933

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Hideo Taka, hereby declare and say as follows:

That I received a Doctorate of Science from Tokyo
Metropolitan University in 2000.

That since 2003, I have been employed by Konica Corporation, now named as Konica Minolta Holdings Inc., the Assignee of the above-referenced application, and have been engaged in the research and development in the field of organic light-emitting device.

That I am one of the inventors of the present application.

That I am familiar with the subject matter of the present invention.

Experimental

The following experiments were carried out by Hideo Taka, one of the inventors of the present application.

A purposes of the present experiments is as follows:

With respect to claim 3 of the present Application, the Examiner states in pages 4 - 5 of the outstanding Office Action that Takimoto et al. (US 5,331,182 A) disclose a polymer with arylene units Ar_2 (column 8, lines 45-65) and L_2 units O, S, Se, or Te (column 2, line 59), and wherein n is not less than 2 (column 2, line 59), while Takimoto et al. does not explicitly disclose the polymer containing pendent chains attached to Ar_2 , that Tokito et al. (US 2003/0091862 A1) teach attaching pendent chains of hole transport compounds [0090], such as carbazole ([0090] HT-1) and

phenylcarbazole ([0095], first side chain in polymer P3), and phosphorescent complexes [0106] to the polymer backbone, and that it would be obvious to one of ordinary skill in the art at the time of the invention to combine the hole transport compounds and phosphorescent complexes pendent chains as taught by Tokito et al. with the polymer of Takimoto et al.

The Examiner also states in pages 6 - 7 of the outstanding Office Action that Ihara et al. (US 2002/0193532) teach attaching pendent chains of phosphorescent complexes to the polymer backbone [0044], the phosphorescent organometallic complexes having partial structures of instant formulae (5) and (7) [0044], and that it would be obvious to one of ordinary skill in the art at the time of the invention to combine the phosphorescent complexes as taught by Ikehira et al. with the polymer of Takimoto et al.

However, in Tables 5 (page 106) and 7 (page 107) of the present Application, it is clearly demonstrated that, when a carbazole group (a hole transport group) is attached to the main chain of the polymer represented by Formula (2) of the present Application as a pendent chain, namely, attached to Ar₂ of Formula (2) (Organic EL Element 2-1-7 in Tables 5 and 7), unexpectedly excellent properties were obtained compared to when the same carbazole group is attached to the main

chain of a vinyl polymer as a pendent chain (Organic EL Element 2-1-1 in Tables 5 and 7) which is also disclosed by Tokito et al. ([0095], first side chain in polymer P1). Such unexpected result can never be obvious even over the combination of Takimoto et al. and Tokito et al.

A purpose of the present experiments is to provide further examples of polymers exhibiting unexpected properties when incorporated in an organic EL element to show the non-obviousness of the polymers containing repeating units represented by Formula (2) of the present Application.

In the present experiments, organic EL element properties of each of the following combinations of compounds were compared (numerals in the parenthesis each represent Organic EL Element number shown in the following Tables):

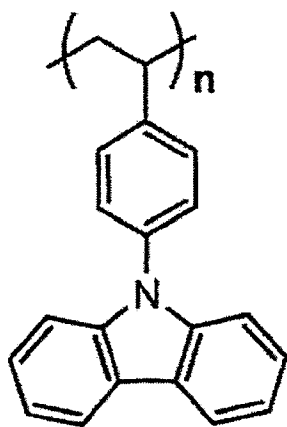
Combination 1: PO-11/Ir-1 of the present Application
(2-1-14) vs. PSCz/Ir-1 (2-1-18)

Combination 2: PO-12/Ir-1 of the present Application
(2-1-15) vs. PSCz/Ir-1 (2-1-18)

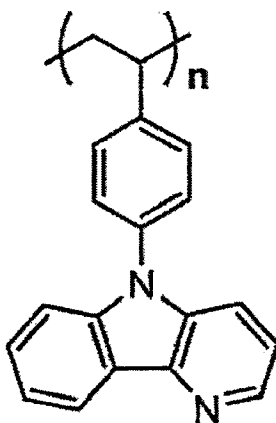
Combination 3: PO-13/Ir-1 of the present Application
(2-1-17) vs. PSaCz/Ir-1 (2-1-19)

Combination 4: Poly-43/Ir-1 of the present
Application (2-3-1) vs. PVETPBI/Ir-1 (2-3-2)

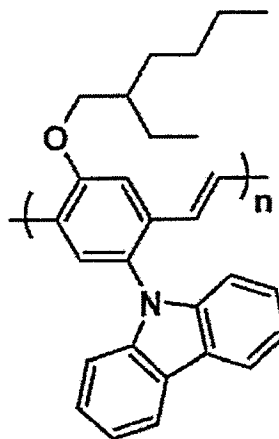
Combination 5: Poly-17/Ir-1 of the present
 Application (2-4-1) vs. CzPPV/Ir-1 (2-4-2)



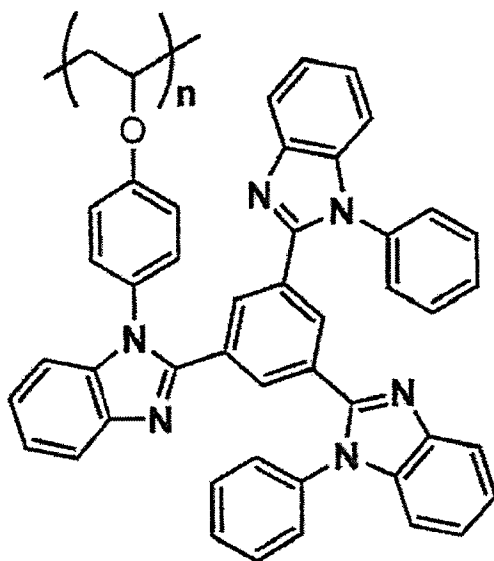
PSCz



PSaCz



CzPPV



PVETPBI

<Preparation of organic EL elements>

A pattern was formed on a substrate (100 mm x 100 mm x 1.1 mm) composed of a glass plate and a 100 nm ITO (indium tin oxide) layer (NA45 manufactured by NH Technoglass Co., Ltd.) as an anode. Then the resulting transparent substrate having the ITO transparent electrode was subjected to ultrasonic washing in i-propyl alcohol and dried by a dry nitrogen gas and subjected to UV-ozone cleaning for 5 minutes. 30 mg of polyvinylcarbazole (PVC) and 1.8 mg of Ir-1 were dissolved in 1 ml of dichlorobenzene and spin coated on thus obtained transparent substrate at 1000 rpm for 5 seconds (thickness: around 100 nm), followed by vacuum drying at 60°C for 1 hour to form a light emitting layer.

The substrate was fixed in a vacuum deposition apparatus and the pressure in the vacuum tank was reduced to 4×10^{-4} Pa. Then, a 0.5 nm thick lithium fluoride layer as a cathode buffer layer and a 110 nm thick aluminum layer as the cathode were deposited, followed by sealing with glass. Thus, Organic EL element 2-1-1 was prepared.

Organic EL elements 2-1-17, 2-1-18, 2-1-19, 2-3-1, 2-3-2, 2-4-1 and 2-4-2 were prepared in the same manner as organic EL element No. 2-1-1, except that PVK and Ir-1 used in the light emitting layer were changed to the compounds listed in Tables 5-continued, 7-continued and 9 - 12.

<Evaluation of organic EL elements>

On the above organic EL elements, the following evaluation was carried out.

(External Quantum Efficiency)

Electric current of 2.5 mA/cm^2 was supplied to each sample at 23° C in an atmosphere of a dry nitrogen gas, and the external quantum efficiency (%) of each sample was measured. The external quantum efficiency (%) was calculated from the data obtained by being measured through a spectral radiance meter CS-1000 produced by Konica Minolta Sensing Inc.

(Emission life)

Electric current of 2.5 mA/cm^2 was supplied to each sample at 23° C in an atmosphere of a dry nitrogen gas, and measured was the duration in which the luminance of each sample decreased to half of the initial luminance, which was designated as the half life of emission ($\tau_{0.5}$) and used as an index of emission life. The measurement was carried out using a spectral radiance meter CS-1000 produced by Konica Minolta Sensing Inc.

(Driving voltage)

The voltage at which emission started was measured at 23° C in an atmosphere of a dry nitrogen gas. The voltage at

which emission started was determined by measuring the voltage at which the luminance increased to 50 cd/m². The measurement of luminance was carried out using a spectral radiance meter CS-1000 produced by Konica Minolta Sensing Inc.

The values of external quantum efficiency, emission life, and driving voltage for organic EL elements 2-1-17 to 2-1-19 were listed in Table 7-continued as relative values when each value of organic EL element 2-1-1 was set to 100.

The values of external quantum efficiency, emission life, and driving voltage for organic EL element 2-3-1 were listed in Table 10 as relative values when each value of organic EL element 2-3-2 was set to 100.

The values of external quantum efficiency, emission life, and driving voltage for organic EL element 2-4-1 were listed in Table 12 as relative values when each value of organic EL element 2-4-2 was set to 100.

Results and Discussion

The obtained organic EL element properties were summarized in following Tables 5-continued, 7-continued and 9 - 12. In tables 5-continued and 7-continued, parts of the data contained in original Tables 5 (page 106) and 7 (page

107) of the present Application were also listed for easier comparison of the data.

Table 5-continued

| Organic EL Element | Light emitting layer | Number average molecular weight of polymer | Remarks |
|--------------------|----------------------|--|-------------|
| 2-1-1 | PVK/Ir-1 | 110,000 | Comparative |
| 2-1-14 | PO-11/Ir-1 | 25,000 | Inventive |
| 2-1-15 | PO-12/Ir-1 | 19,000 | Inventive |
| 2-1-17 | PO-13/Ir-1 | 21,000 | Inventive |
| 2-1-18 | PSCz/Ir-1 | 67,000 | Comparative |
| 2-1-19 | PSaCz/Ir-1 | 23,000 | Comparative |

Table 7-continued

| Organic EL element | External quantum efficiency (Relative value) | Emission Life (Relative value) | Driving voltage (Relative value) | Remarks |
|--------------------|--|--------------------------------|----------------------------------|-------------|
| 2-1-1 | 100 | 100 | 100 | Comparative |
| 2-1-14 | 231 | 662 | 76 | Inventive |
| 2-1-15 | 237 | 901 | 79 | Inventive |
| 2-1-17 | 234 | 780 | 75 | Inventive |
| 2-1-18 | 96 | 101 | 106 | Comparative |
| 2-1-19 | 94 | 95 | 105 | Comparative |

Table 9

| Organic EL Element | Light emitting layer | Number average molecular weight of polymer | Remarks |
|--------------------|----------------------|--|-------------|
| 2-3-1 | Poly-43/Ir-1 | 12,000 | Inventive |
| 2-3-2 | PVETPBI/Ir-1 | 18,000 | Comparative |

Table 10

| Organic EL element | External quantum efficiency (Relative value) | Emission Life (Relative value) | Driving voltage (Relative value) | Remarks |
|--------------------|--|--------------------------------|----------------------------------|-------------|
| 2-3-1 | 190 | 545 | 82 | Inventive |
| 2-3-2 | 100 | 100 | 100 | Comparative |

Table 11

| Organic EL Element | Light emitting layer | Number average molecular weight of polymer | Remarks |
|--------------------|----------------------|--|-------------|
| 2-4-1 | Poly-17/Ir-1 | 13,000 | Inventive |
| 2-4-2 | CzPPV/Ir-1 | 12,000 | Comparative |

Table 12

| Organic EL element | External quantum efficiency (Relative value) | Emission Life (Relative value) | Driving voltage (Relative value) | Remarks |
|--------------------|--|--------------------------------|----------------------------------|-------------|
| 2-4-1 | 1500 | 108 | 96 | Inventive |
| 2-4-2 | 100 | 100 | 100 | Comparative |

From the data listed in Tables 5-continued, 7-continued and 9 - 12, the following results were obtained:

With respect to above Combinations 1 and 2, Organic EL elements 2-1-14 and 2-1-15 each containing a polymer having a repeat unit in which a phenyl carbazol group exhibiting a hole transport property (refer to page 33, HT-5) is pendent to a polymer chain of the present Application showed an unexpectedly higher external quantum efficiency, an unexpectedly longer emission life and an unexpectedly lower

driving voltage, when compared with Organic EL elements 2-1-18 containing a polymer having a repeat unit in which the same phenyl carbazol group is pendent to a vinyl polymer chain.

With respect to above Combination 3, Organic EL element 2-1-17 containing a polymer having a repeat unit in which a phenyl carbolinyl group exhibiting an electron transport property (refer to page 41, ET-13) is pendent to a polymer chain of the present Application showed an unexpectedly higher external quantum efficiency, an unexpectedly longer emission life and an unexpectedly lower driving voltage, when compared with Organic EL elements 2-1-19 containing a polymer having a repeat unit in which the same phenyl carbolinyl group is pendent to a vinyl polymer chain.

With respect to above Combination 4, Organic EL element 2-3-1 containing a polymer having a repeat unit in which a group exhibiting an electron transport property (known as TPBI) is pendent to a polymer chain of the present Application showed an unexpectedly higher external quantum efficiency, an unexpectedly longer emission life and an unexpectedly lower driving voltage, when compared with Organic EL elements 2-3-2 containing a polymer having a

repeat unit in which the same electron transport (TPBI) group pendent to a vinyl polymer chain.

With respect to above Combination 5, Organic EL element 2-4-1 containing a polymer having a repeat unit in which a carbazolyl group exhibiting a hole transport property (refer to page 33, HT-1) is pendent to a polymer chain of the present Application showed an extremely higher external quantum efficiency, a longer emission life and a lower driving voltage, when compared with Organic EL elements 2-4-2 containing a polymer having a repeat unit in which the same carbazolyl group is pendent to a conjugated polymer chain.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001, of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Dated: November 24, 2009

Hideo Taka

Hideo Taka